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# NANOSTRUCTURES AND ENHANCED PROPERTIES IN TUNGSTEN AND ITS ALLOYS PROCESSED BY EQUAL CHANNEL ANGULAR PRESSING

**Final Technical Report**


**by**

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**13. ABSTRACT** The aim of the present project has been the development of the ECAP technique for processing of UFG structures in tungsten (W) and W-4.3%Ni-2%Fe-alloy, which refer to very low-ductility and hard-to-deform materials. Taking into account the computer simulation results of ECAP there has been conducted a modernization of the die-set, aimed at decreasing the pressing temperature below 1000 °C at the cost of a decrease in the intensity of deformation for one pass and a use of backpressure. The influence of the number of passes, the rate and the pressing route on the UFG structure formation has been investigated. There has been shown the possibility to obtain the UFG structure with a grain size about 0.5 μm in W by optimizing the ECAP regimes. A formation of such small grains leads to a strength enhancement. At the same time manifestations of the viscous character of the UFG W fracture points out at a principal change in its ductile properties. A strong microstructure refinement after several passes of ECAP has been achieved also in the W heavy alloys with a low workability.

There have been outlined ways for a further progress in ECAP of W, leading to a formation of a homogeneous UFG structure in bulk billets, which provides extraordinary mechanical properties.

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## ABSTRACT

The aim of the present project has been the development of the equal-channel angular pressing (ECAP) technique for processing of ultrafine grained (UFG) structures in commercially pure tungsten (W) and its alloy W-4.3%Ni-2%Fe, which refer to low-ductility and hard-to-deform materials. Taking into account the computer simulation results of ECAP we have conducted a modification of the die-set, aimed at decreasing the pressing temperature below 1000 °C at the cost of a decrease in the intensity for one pass of deformation and the use of backpressure. The influence of the number of passes, the rate and the pressing route on the UFG structure formation has been investigated. There has been shown the possibility to obtain the UFG structure with a grain size about 0.5 μm in W by optimizing the ECAP regimes in the frames of this work. A formation of such small grains leads to strength enhancement. At the same time the manifestations of the tough character of W fracture points out to a principal change in its ductile properties.

A strong microstructure refinement after several passes of ECAP has been achieved also in W heavy alloys with a low workability.

There have been outlined ways for a further progress in ECAP of W, leading to a formation of a homogeneous UFG structure in bulk billets, which provides extraordinary mechanical properties.



**Keywords:** *severe plastic deformation, ultrafine-grained materials, W and its alloys, equal-channel angular pressing, microstructure refinement, microhardness, mechanical properties.*


## 1. INTRODUCTION

A formation of ultrafine-grained nanostructures in metallic materials by the evere plastic deformation (SPD) techniques provides a potential to achieve their novel and extraordinary properties [1-3]. Equal-channel angular pressing (ECAP) appears to be one of the most successful and commercially available methods to obtain bulk UFG structures in different metals and alloys. A significant progress achieved in regulating the microstructure and the properties of ductile materials has made it possible to start investigations concerning the microstructure refinement and the improvement of mechanical properties in low-ductility and hard-to-deform materials.

Within the frames of our previous project № 68171-99-M-6634 "Developing nanostructures and enhanced properties in heavy W alloy through SPD processing" we demonstrated that unlike conventional methods of metal-working, such as extrusion and rolling, ECAP allowed to produce the UFG structure with an average grain size of about 1 μm in bulk billets of commercially pure W. At the same time, numerous investigations testify, that considerable enhancement of mechanical properties, as a result of nanostructure formation is a complex problem, which depends on the multiple processing and microstructure parameters.

The current report presents the results of experimental research and numerical simulation related to the designing, controlling and optimising the ECAP processing, which ensure producing of bulk W billets with an average grain size less than 1 μm and as a result the enhancing of mechanical properties.

## 2. MATERIAL AND EXPERIMENTAL PROCEDURES

The investigation has been performed on commercially available W in the shape of rods 15 mm in diameter, manufactured by powder metallurgy methods. The rod has been produced by means of hot rolling and rotation forging of the sintered powder. The impurities content of the as-received materials has been (% to mass): O - 0.0094, N - 0.00066, C - 0.0134 (the measurement error for oxygen and nitrogen is 1.0%, for carbon - 0.5% of the measured value).

The heavy W-4.3%Ni-2%Fe-alloy has been delivered for the investigations as a turned rod 17.5 mm in diameter and 300 mm in length.

The ECAP process has been carried out using a special die-set heated up to 500 $^0$C on the samples 15 mm in diameter and 60 mm in length. In order to decrease the oxidation of the metal during heating and to reduce the friction coefficient, W billets before pressing were placed into a cylindrical steel shell.

High pressure torsion (HPT) straining has been performed using Bridgman anvils pre-heated up to 500 °C under the imposed pressure of about 6 GPa on electropolished samples in the shape of a disc 10 mm in diameter and 0.8 mm in thickness.

Microstructural investigations of ECAP billets have been performed in transverse and longitudinal sections by using the optical microscopy (OM) (NEOPHOT-32), the scanning electron microscopy (SEM) and the transmission electron microscopy (TEM) (JEM-100). For the OM, chemical etching of the polished metallographic samples in a 3% solution of hydrogen peroxide has been used. These W foils have been prepared by jet-polishing in the water solution of sodium hydroxide at 10 °C and 10 V and the solution of 5% $H_2SO_4$ and 2% HF in methanol at -40 °C has been used to prepare foils of the W heavy alloy. A mean grain size and a volume fraction of ultrafine grains have been determined by the linear intercept method.

X-ray structural analysis has been carried out on the DRON-4-07 diffractometer. The accuracy of the measurement of the diffraction angles was 0.005°. The values of coherent scattering domain size and elastic

microdistortions in the crystal lattice of the samples under study were determined by the method of harmonic analysis using pairs of X-ray peaks (110) and (220). A relative error of the measurement of these values did not exceed 2%.

Mechanical properties were studied by microhardness measurements and tensile tests. Microhardness measurements were made by using the Vickers' method in transverse and longitudinal sections of the polished samples under a load of 200 g applied for 15 s. Each data point represents the averaging out of 10 measurements. Tensile samples having gauge dimensions 3x2x15 $mm^3$ were cut out by spark erosion from pressing billets in longitudinal directions. Tensile test were conducted in the air at a temperature interval between 100-600°C, and at the initial strain rate $1x10^{-3}$ $c^{-1}$ using screw-driven Instron machine equipped with a radiant furnace.

## 3. SPD PROCESSING

A formation of UFG structures during SPD depends on various processing parameters such as the true strain, the temperature and strain rate, the imposed pressure, lubrication, etc. [1]. The establishing and clarifying of the influence of these factors are important in the die-set designing, controlling and optimising the processing conditions, aimed to produce homogeneous UFG structures with the smallest possible grain size.

For producing of UFG structures, ECAP is to be performed as multiple pressing of a bulk billet through two identical channels intersecting at a certain angle Φ. The principle of ECAP is illustrated schematically in Fig. 1. A value of true strain introduced in the billet during ECAP is a function of the value of the angle Φ and the number of passes through the die. For Φ=90° the total accumulated strain is approximately equal to the number of passes [4]. It has been shown that from the grain refinement point of view the angle of Φ=90° and $B_c$ (rotation of the sample by 90° between passes) are the most effective ones [4].

The majority of the results on ECAP were obtained recently in relatively ductile metals and alloys with FCC-structure. The microstructure development of these materials during ECAP can be described on the basis of the concept of cell formation and evolution [2]. Plastic deformation occurs by nucleation and a movement of dislocations. These dislocations tend to rearrange themselves so that dislocation cells appear. Upon straining, new dislocations are accumulated and misorientation angles are increased, leading to a reduction in the cell size. Cell structures are formed in metals and alloys with a high symmetry, high stacking fault energy and an easy cross-slip.

Further of the structure formation for metals and alloys with BCC-structure in comparison with FCC-metals are determined by a drastic temperature dependence of the flow stress, which is originated by the impurities - C, N, etc. – leading to a decrease in ductility and the Peierls-Nabarro high stresses for the dislocation moving. It has been shown in [6] on the basis of the analysis of dislocation structure evolution's investigations of several BCC-metals (chromium, molybdenum and vanadium) in a wide range of temperatures and strain rates, that equiaxed misoriented cell structure formation is observed in the area of temperatures high enough, but somewhat lower than the temperature of recrystallization. The recrystallization temperature of W is about 1400 °C. A decrease by 200-300 °C is possible after large deformations. Thus, in order to form the UFG structure the ECAP temperature of W and its alloys should not exceed 1000-1100 °C. However in such conditions these metals have a low workability, due to a low ductility and the enhanced strength. Besides, an intensive oxidation of W takes place at high temperatures leading to the enhanced failureability. That is why ECAP of W and its alloys has been conducted using a covering shell. Since W has high thermal conductivity and low thermal capacity, the shell, apart from the oxidation protection, should have also decreased heat losses and improved tribological conditions during ECAP.

The friction between the tool and the billet is an important parameter, which influences the homogeneity of plastic metal flow during ECAP [12]. Additional insight to the effects of friction conditions on the deformation flow has been fulfilled in the present work through the finite-element simulation (FEM) method. Numerical simulation for the material with isotropic hardening showed that with a decrease in the friction coefficient from 0.2 to 0 the homogeneity of the plastic flow is increasing (Fig. 2) [19].

Thus, all this demonstrates that ECAP of W and its alloys is a complex task requiring an investigation of the scientific and technical problems identified above.

According to our last year report [22], a special die-set was designed and manufactured to process W billets without failures. Since W has a low ductility at temperatures close to 1000 °C, the angle of channel intersection in the die has been increased for this work (an angle of 90° is typically used for ductile materials). The determined solution has been confirmed by attempts to conduct ECAP with an angle of channels intersection of 90°. In this case billets have failed on the first or second pass [22]. With a change in the angle of channel intersection from 90° to 110° a shear strain value for one pass of the ECAP process of W has been reduced from e=1.15 to e=0.8 and the total accumulated strain value for 8 passes has been =6.4.

While developing the ECAP for W at approximately 1000 °C two routes with different billet rotation (namely route $B_c$ and route C) have been selected. These routes provide usually a formation of equiaxed structures in metals [8]. However our investigations have shown that only route C can provide billet processing without failure in case of CP W [22]. This is attributed to the fact that its necessary workability can be realized at processing by the route C, but not by the route B. Considering the positive influence of a low strain rate on the workability and homogeneity of the material flow [11], the ECAP has been conducted at a minimum strain rate at which the equipment could be used; namely 6 mm/s. Steel 2 mm in thickness has been used as a covering shell.

It is shown in our works [21] that the UFG structure with a mean grain size about 1 μm is being formed as a result of ECAP on the above-mentioned regimes in CP W. Whereas, ECAP of Cu, Al and Ni can provide the grain size equal to 0.2-0.3 μm [2].

A minimal grain size depends to a great extent on the stacking fault energy (SFE). In the general case with an increase in SFE the grain size is increasing as well [2]. The more the value of SFE, the sooner (according to the stress level and the amount of strain) the structural reorganization, leading to a cell structure formation, takes place in the material. In metals and alloys with BCC-lattice at similar temperatures reorganization of the dislocation structure occurs ten times faster, than in FCC-metals, because diffusion and dislocation recovery are sufficiently higher. As a result it leads to a decrease in dislocation accumulation rates as well as to a decrease in the hardening rate in BCC-metals and therefore it causes a formation of cells, having a larger size at the same straining.

A further refinement of a grain size can be achieved by decreasing the deformation temperature or increasing the intensity of the ECAP processing. At the same time, as it has been mentioned above, the temperature decreasing or increasing of a strain intensity results in the sample failure due to a lack of its workability. This problem can be solved by an increase in the angle of channels intersection and in the number of passes, using backpressure during ECAP.

A principal possibility to achieve large deformations at low temperatures in the conditions, close to hydrostatic compression has been demonstrated at severe plastic deformation of CP W by high-pressure torsion (HPT). An application of HPT straining at T=500 °C under the imposed pressure of about 6 GPa enables to process disc-shaped samples with the true logarithmic strain equal to 10 without any microcracks [21].

An installation of the ECAP die-set on the crank press has allowed increasing the pressing rate up to 300 mm/s. An experimental research of CP W subjected to ECAP with an intersecting angle equal to 120° showed that an increase in the speed of ECA pressing from 6 to 300 mm/s enabled to decrease the heating temperature of the billet from 1000 °C to 950 °C and to produce a bulk billet after 6 pressing passes without any cracks.

At the same time, as it has been mentioned above, the decreasing or increasing temperature of the accumulated strain results in the sample failure due to a lack of its ductility. This problem can be solved using backpressure during ECAP process. In order to produce a necessary level of backpressure, the length of the exit channel has been increased. The results of the investigation showed that an increase in the length of the exit channel up to 35 mm led to a working pressure increase from 1000 to 1300 MPa. However, microcracks were revealed in W samples after 10 passes under the speed 300 mm/s and the die angle 120° of ECA pressing at 950 °C, which indicates the necessity of the back pressure value increase. In that case the protection steel shell was not sufficiently hard which led to an irregular deformation of the initial cylindrical W billet after a high speed ECA pressing. Consequently, after large cycles of processing the billet had a cross-section of a variable size.

The presented results show, that the process of ECAP is complex and one needs to find optimal processing routes and regimes to obtain a homogeneous UFG-structure and the enhanced properties. In this connection, the analysis of ECAP by FEM approaches may provide useful insights into the effect of the die-geometry of the metal flow, the stress-strain state and contact stress. It is useful for the tool designing and the processing modifying.

In the present work, continuing our previous research [19] the plastic deformation behaviour of the material during multi-passes ECAP by two routes A and C has been studied using the 2D deformation simulations. For the numerical computation of the plastic flow a material model with isotropic hardening has been applied. The finite elements mesh system for the plastic flow of W simulation was used in this investigation. The finite element model of the process has been consisted of the mesh of 285 elements and 500 nodes.

The results of the plastic flow computation are represented as isolines and isostrips of different stress-strain components. The equal values isolines of the accumulated plastic values, rate intensity and stresses during the billet passing through the tool channels at a consecutive realization of two passes on routes A and C are represented in Fig. 3 and 4 respectively.

A comparative analysis of two flow simulation regimes has allowed revealing the following peculiarities:

1. In both of the regimes we observed the heterogeneous flow with a formation of local maxima of the stress intensity. At the same time a more homogeneous stress distribution is characteristic of the route A, and the formation of the largest stresses is characteristic of the second pass.

2. The analysis of shear stresses has shown, that the route A has a more homogeneous shear stress distribution, than the route C. Meanwhile, maximal values of the shearing stresses are observed at the plastic flow on the route C.

3. The analysis of plastic strain distribution makes it possible to conclude that the route A leads to a processing of a more homogeneous strain distribution, than the route C.

4. The study of the accumulated deformation when changing an ECAP route has shown, that the value of the accumulated strain on the route A reaches 0.8953. A rapid accumulation of the plastic deformation on the route C determines higher values of the accumulated strain 1.163.

The simulation results obtained led to a better understanding of the mechanics of ECAP, which were very useful for the die-set design and optimisation of process conditions.

By applying the results of the numerical simulation and the experimental research of ECAP process, a modernization of the die-set, aimed at decreasing the pressing temperature below 1000 °C at the cost of a decrease in the deformation intensity and an application of back pressure equal 2.4 of W yield stress at 900 °C have been performed. An increase in the angle of channels intersection up to 135° leads to a shear strain value for one ECAP pass reducing to $e \approx 0.5$. In order to produce a high level of backpressure up to 1300 MPa, the diameter of the exit

channel has been decreased from 16 mm to 14.5 mm. We used also an installation of the ECAP die-set on the crank press, which allowed increasing the pressing rate up to 300 mm/s.

Modernization of the die set and optimisation of strain rates conditions have enabled to produce at 900 °C bulk W billets with the total accumulated strain value after 16 passes (e ≈8) without any cracks (Fig. 5).

The W heavy alloy exhibits a unique combination of high strength, high density and toughness [17]. However, first experiments on ECAP of the W-4.9%Ni-2.1%Fe-heavy alloy have shown a number of processing problems. A low workability of this alloy makes the billets fail even after 1-2 passes at a temperature of 1000 °C and with an angle of channels intersection equal to 120°. A temperature increase up to 1100-1150 °C has not also led to a successful ECAP with several passes. In this connection a special die with an angle of channels intersection equal to 135° and the backpressure has been fabricated to decrease the strain intensity (Fig. 6). The die modification has allowed the heavy alloy billets samples not to fail after the forth passes of ECAP (Fig. 7). However the next pass has resulted in the billet failure.

According to [20] the microstructure and properties of W heavy alloys are sensitive to many factors including the purity of raw materials, sintering conditions (temperature, time, atmosphere, cooling rate) of the initial billets, heat treatment parameters, W content and the rate of plastic strain. In this connection a further work should be presented by complex investigations of factors influencing the workability of heavy W alloys at high temperatures and ECAP should be conducted in conditions, close to the isothermal ones with a possibility to control strain behaviour of the billets at ECAP. With this aim to our mind the special die set with an angle of channel intersection 120° from the heat-resistant Ni -based superalloy enabling to perform ECAP up to the temperature of 900 °C should be fabricated for a successful processing of W billets.

## 4. Microstructure refinement

### 4.1. Commercially pure tungsten

The structure of the as-received W is composed of large, almost equiaxed grains with a diameter of 60-80 μm in transverse section of the initial rod (Fig. 8). In the longitudinal section of the rod, grains are elongated along its axis with an aspect ratio about 10. TEM studies show the presence of a well-formed polygonized dislocation substructure inside of coarse grains. This substructure has a low density of the lattice distribution.

We showed in our previous report [22] that ECAP of W conducted at 1050 °C using the die-set with Φ=110° by the route C, i.e. with a rotation of a billet between subsequent passes through 180° around its longitudinal axis, led to a significant microstructure refinement, the character of which substantially depends on the number of passes.

From the optical microstructure of W after 4 passes of ECAP shown in Fig. 9 it can be seen clearly that 4 passes lead to development of smaller grains as compared to the initial state. An average grain size is less than 20 μm in transverse section (Fig. 9, a). In the longitudinal section the grain structure of the billets remains elongated.

An increase in the strain up to 8 passes has resulted in a further grain refinement as well as in an increase of its homogeneity. As can be seen from Fig. 9, after such a processing, there occurs a formation of a rather ultrafine-grained structure, equiaxed and sufficiently homogeneous both in the transverse and the longitudinal sections of a billet (Fig.9, c, d). The mean grain size in this ECAP state cannot be estimated precisely by the OM observation, because of too small grains.

In order to clarify microstructure changes in the ECAP W, TEM examination and selected area electron diffraction (SAED) have been applied. The changes in TEM micrographs of W with an increase in the number of passes of ECAP are shown in Fig. 10. TEM studies testify that the initial polygonized substructure is completely transformed to a fine structure of subgrains and grains. After 4 passes a well-refined substructure is being formed (Fig. 10, a, b). The given substructure differs significantly from the substructure in the initial W. There are mixed subgrains of different shape and sizes, with and without high dislocations density, which are located in tangles and dislocation cell walls. Some subgrain boundaries possess the high angle misorientation. This is confirmed by the selected area diffraction pattern in which one can see the appearance of a number of spots arranged in a circle. As the number of passes increases from 4 up to 8, subgrains become finer with sharper boundaries (Fig. 10 c, d). Note should be made that many of the subgrains are equiaxed and dislocation-free. Spreading of numerous spots also indicates the presence of high internal elastic stresses in the microstructure. The mean grain size is about 1μm, though in the structure one can see an appearance of the number of finer grains, 0.3-0.5 μm in size.

Further grain refinement can be achieved by decreasing the deformation temperature and/or increasing the strain. To investigate a possibility of maximal grain refinement in W using severe plastic deformation we have performed its HPT straining. Fig. 11 illustrates the effect of HPT with 5 rotations at 500 °C under the imposed pressure equal to 6 GPa. It can be seen that severe deformation by HPT has resulted in a strong microstructural refinement up to a mean grain size measured from the dark field image of about 100-200 nm (Fig. 11, b).

As it can be seen also from the SAED pattern, the majority of new grains have random misorientations. The bent extinction contours within the crystallites indicate the presence of high internal stresses due to high density of dislocations and in the vicinity of boundaries.

In this connection for a further grain refinement during our ECAP experiments the deformation temperature has been decreased from 1050 to 900 °C and the angle of channels intersection Φ has been increased from 110° to 135°. It is known [18] that a decrease in the strain intensity per pass during ECAP may strongly

affect the microstructure refinement. Fig. 12 illustrates the effect of the number of passes through the 135 ° die on the microstructure developed by ECAP process. After 8 passes, which corresponds to a total strain close to 4 the microstructure is strongly refined, but it is rather heterogeneous and new grains are elongated especially on the longitudinal section (Fig. 12b). With further straining up to e≈8 after 16 passes, the microstructure refinement is accompanied by an increase in structure uniformity (Fig. 12, c, d). The inspection of the selected area diffraction pattern containing many spots has allowed suggesting that some high angle grain boundaries are appeared in the microstructure of the sample, though the majority of new grains have still the low angle orientation. At the same time the banded shape of (sub)grain structure becomes less evident. The structure of longitudinal section of the processed billets consists of slightly elongated (sub)graines (Fig. 12, d).

X-ray structural analysis revealed a significant broadening of diffraction peaks resulting from microstructure refinement and high dislocation density. The size of coherent scattering domains and the value of microstrains before and after ECAP and HPT straining are presented in Table 1. As it can be seen from Table 1, the line broadening after HPT straining is associated with a larger elastic microdistorsion than after ECAP processing. At the same time, the crystallite size after HPT straining has become comparative with the TEM grain size, which does not occur in ECA pressed W.

The gap between the microstructures produced by those two SPD techniques has a strong effort on the material hardness. The microhardness after HPT straining is almost twice as large as the microhardness in the as-received state and is equal to 10.5 GPa. However, the maximal microhardness for different processing routes is not higher than 8-9 GPa after ECAP.

### 4.2. The tungsten heavy alloy

The investigated W heavy alloy is a two-phase material composed of a harder W grains and a softer matrix. As it is shown in Fig. 13 (a, b), the spherical W grains about of 25-30 μm in diameter are quite homogeneously distributed in the matrix in both transverse and longitudinal sections. However, after 4 passes by ECAP processing using the 135° die at 1100 °C the W grains become ellipsoidal in the longitudinal section. They are aligned along the direction about 60° from the ECAP direction (Fig. 13, d).

The OM observations revealed also the fragmentation and breaking of some W grains during ECAP processing. The evidence of a subgrain formation in the W grains can be well observed by slight etching (Fig. 13, c). One can see the signs of the intense deformation in the matrix as well, where a formation of some shear bands takes place.

The TEM observations have revealed heterogeneous distribution of lattice dislocations in microstructure of the as-received W heavy alloy (Fig. 14). The low dislocation density is about of $10^6$ $cm^{-2}$ in the harder W grains (Fig. 14a), while the softer matrix contains a significantly higher dislocation density ($5x10^9$ $cm^{-2}$), which are homogeneously distributed in the matrix (Fig. 14, b). The ECAP processing of the W heavy alloy has led to a significant change of its dislocation structure. Different types of dislocation configuration are observed in the W grain and the matrix. An increase in the dislocation density of the W grains is accompanied by a rearrangement of dislocations into cell and/or subgrains. The homogeneous near equiaxed subgrains structure with the size of 0.5-1 μm is developed in the cross section of the billet (Fig. 14, c). The sublongitudinal section of the processed billets consists mainly of the banded substructure of W grains, the width of which is about of 1 μm (Fig. 14, d). In the matrix the subgrain formation has not been detected. Instead of it we can observe an introduction of areas, in which single dislocations are not resolved during ECAP. The main feature of the softer matrix is the formation of the flexural profile of extinction contour, indicating high elastic stresses (Fig. 14, e). This microstructure changes are reflected in a substantial increase in hardness.

## 5. MECHANICAL BEHAVIOUR

The influence of the grain size on the strength ($\sigma_y$ – yeld stresses) or hardness ($H_v$) of metals is commonly expressed by the Hall-Petch equation:

$$\sigma_y=\sigma_o+k_yd^{-1/2} \qquad\qquad H_v=H_{vo}+k_yd^{-1/2}$$

where $k_y$ is the Hall-Petch constant and d is an effective grain size.

A change in the mean grain (subgrain) size measured by TEM with an increase in the amount of strain at ECAP processing of CP W is plotted in Fig. 15 (a). The grain size sharply decreases until strains and after that decreases gradually with an increase in the strain. After 16 passes by the 135 ° die-set with a total strain equal to 8 a mean grain size is about 0.5-0.7 μm.

The dependence of the microhardness on the amount of strain is shown in Fig. 15, b. On the contrary to the change in the grain size, the microhardness gradually increases until strains and after that it increases rapidly with an increase in the strain.

Thus, the data in Fig. 14 (a, b) testify clearly, that the grain refinement at ECAP of W leads to a significant enhancing of its hardness, that follows directly the Hall-Petch equation. Moreover, maximal microhardness values, obtained in W, subjected to ECAP and equal at the present moment to 6.4 GPa, are visibly lower than the value $H_v$, being observed in the case of HPT W, equal to 10.5 GPa. It can be explained by a finer

grain size 0.1-0.2 μm in the latter case. On the other hand, this fact demonstrates the potential of further strength enhancement in W at the cost of a greater grain refinement.

A great interest alongside with the strength enhancement has an increase in ductile characteristics of W and its alloys, which are rather brittle materials at low temperatures. As it is known [11], the ductility of the refractory metals can be enhanced to a great extent at the cost of microstructure refinement. In this connection we have conducted first investigations of mechanical tension properties in UFG W, processed by ECAP, with the grain size equal to 1 μm. These investigations have been carried out in a temperature interval from the room temperature to 600 °C. Their results are shown in Fig. 16. One can see, that a decrease in the deformation temperature leads to a growth of the yield stress and the fracture strength. But the fracture strength is decreased in the area of ductile-brittle transition. It can be seen clearly that the grain refinement up to 1 μm during ECAP led to an increse in the yield stress and fracture strength almost by 50%. The ductile-brittle transition temperature, which is determined at $\sigma_y = \sigma_f$ decrease from 180° in the initial state to 125 °C after ECAP. Obviously, a further decrease in the grains size should lead to grater decrease in the temperature of ductile-brittle transition.

Qualitative changes of the fracture character of the destroyed billets (Fig. 17) also testify to a potential in the ductile properties enhancing of the UFG W. One can see, that, contrary to the transcrystallite brittle fracture of the initial billet (Fig. 17 a), the UFG W has a ductile intercrystallite fracture (Fig. 17, b)

## SUMMARY

Continuing the work, started in our previous annual ERO project #68171-99-M-6634, which revealed for the first time a possibility of a strong grain refinement using equal-channel angular pressing in W and its alloys, the investigations, conducted in the frames of the present project has been aimed to receive in these materials the ultrafine grained structures with a grain size about 0.5 μm and to enhance their mechanical properties.

Taking into account the results of the computer simulation conducted as well as our previous experience, we have carried out the modernization of the die-set for ECAP, connected with increasing the angle between the intersecting channels and also connected with using backpressure. This modernization has allowed decreasing ECAP temperature below 950 °C. Investigations of structure formation in W and the W-4.3%Ni-2%Fe-alloy have been conducted with an application of the given die-set depending on the number of passes and the pressing routes. ECAP regimes, leading to a formation in CP W the UFG structures with a grain size about 0.5 μm have been determined. One can observe a significant enhancement of microhardness, strength and ductile characteristics (a decrease in the temperature of ductile-brittle transitions, an appearance of a ductile fracture) in the samples obtained. At the same time, there is a significant potential of a further enhancement of these properties at the cost of a formation of a more homogeneous UFG structure and an increase in the form of high-angle grain boundaries by further optimizing of the pressing routes. A strong microstructure refinement has been also demonstrated for the W-4.3%Ni-2%Fe-alloy using ECAP. Apparently, the overcoming of the problem of an increase in the number of passes at ECAP of this quite a hard-to-deformed material allows obtaining UFG structure in the given alloy as well. We assume, that the above-mentioned problems of structure formation can be successfully solved at the cost of ECAP parameters optimisation (a choice of backpressure, an increase in the intensity and the rate of deformation and etc.). These investigations may allow demonstrating extraordinary properties in W and, probably in other refractory metals – tantalum, molybdenum, in particular the achievement of high-strength and ductile state even at room temperature that may be a new step in their application.

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# APPENDIX

Fig. 1. Schematic illustration, showing the principle of SPD method by means of ECAP

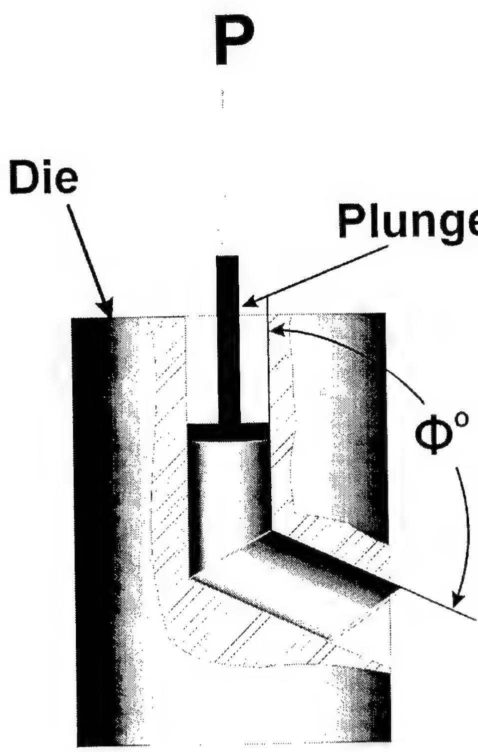



Fig. 2. The FEM distribution of plastic deformation intensity in case of the coefficient k values: (a) k=0.2 and (b) k=0.

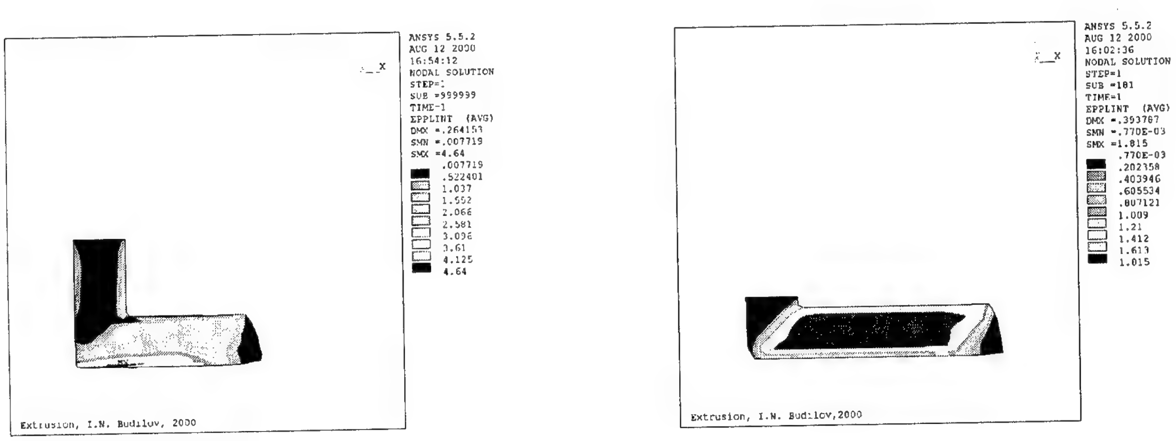


Fig. 3. The FEM results (route A): accumulated strain (a); strain rate intensity (b); strain resistance (c)

Accumulated strain

Strain rate intensity

Strain resistance

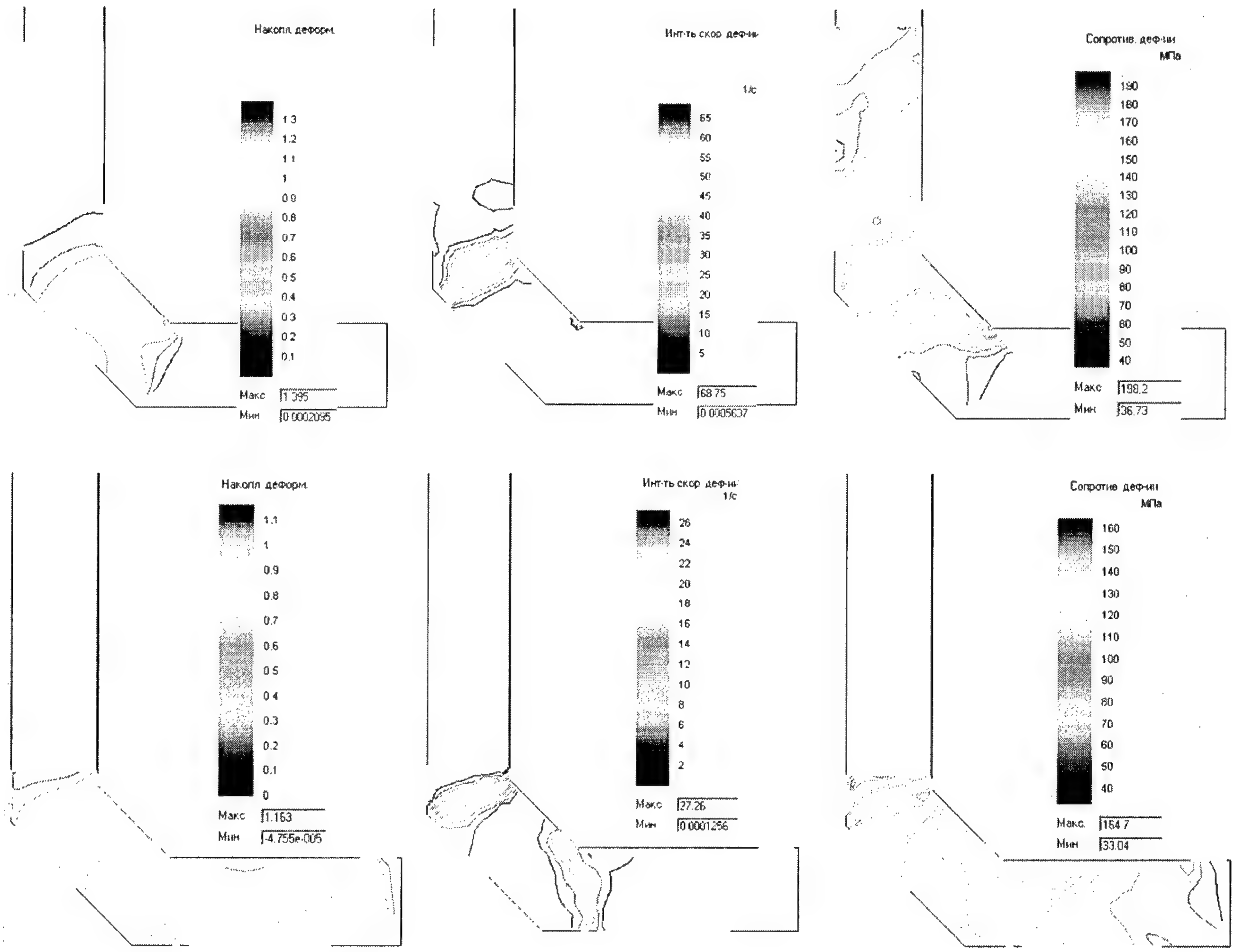


Fig. 4. The FEM results (route C): accumulated strain (a); strain rate intensity (b); strain resistance (c)

Accumulated strain

Strain rate intensity

Strain resistance

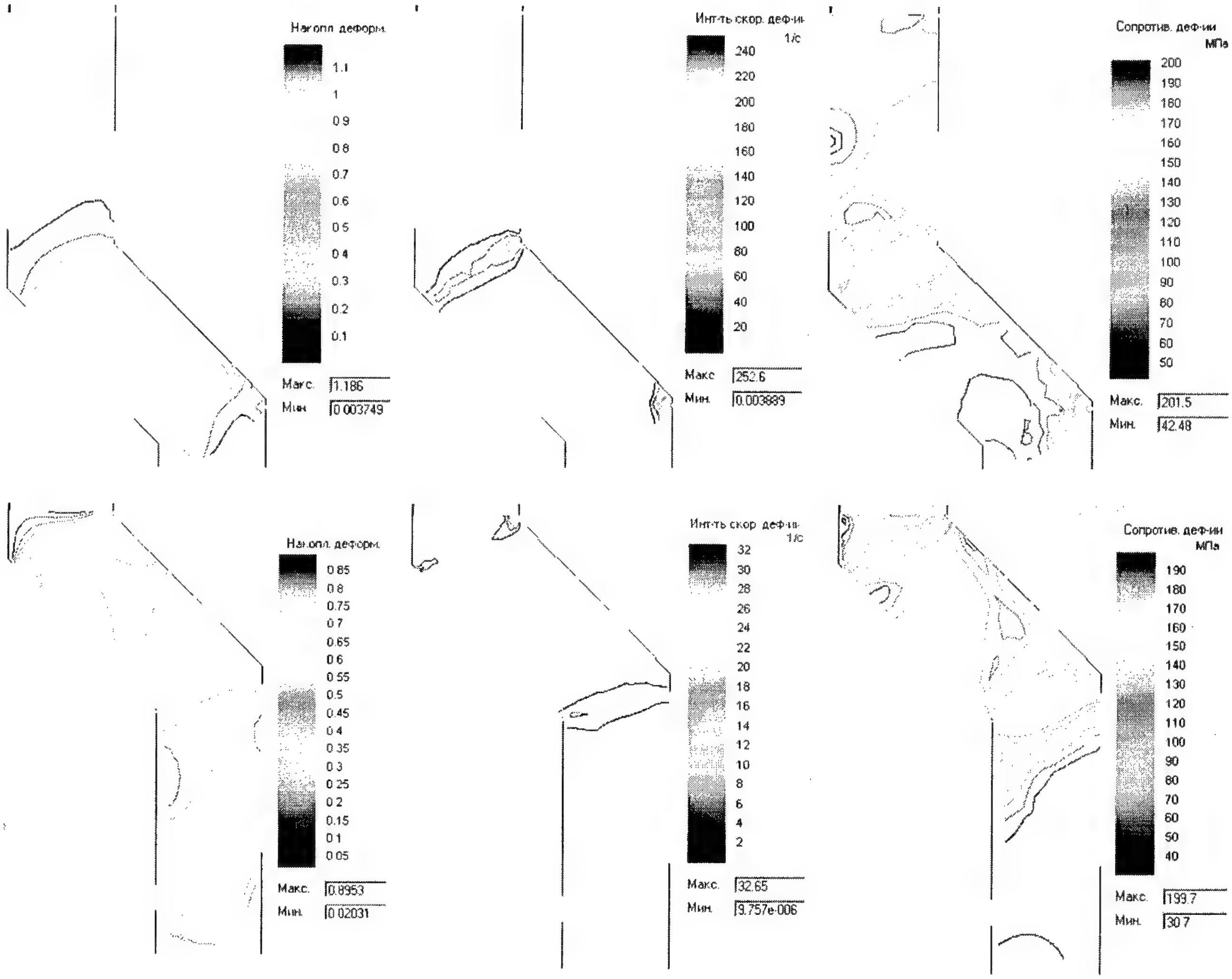



Fig. 5. Bulk W billets before (a) and after (b) ECA pressing by the 135° die at T=900 °C, 16 passes.

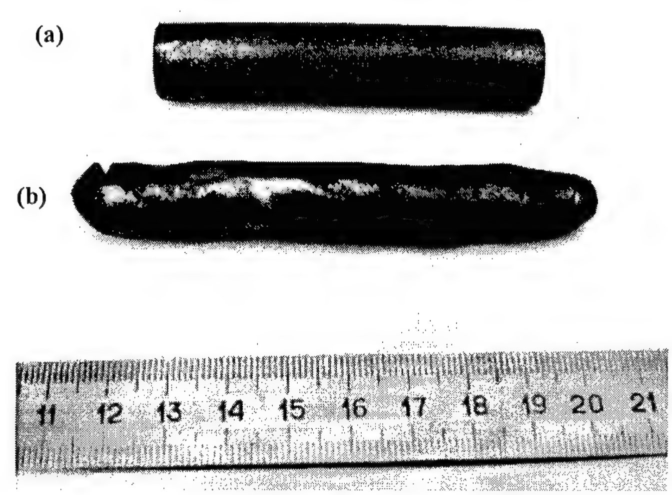


Fig. 6. The special-die set for ECAP of W and its alloys

Fig. 7. Bulk W heavy alloy billets before (a) and after ECA pressing by the 135° die at T=1000 °C, 4 passes

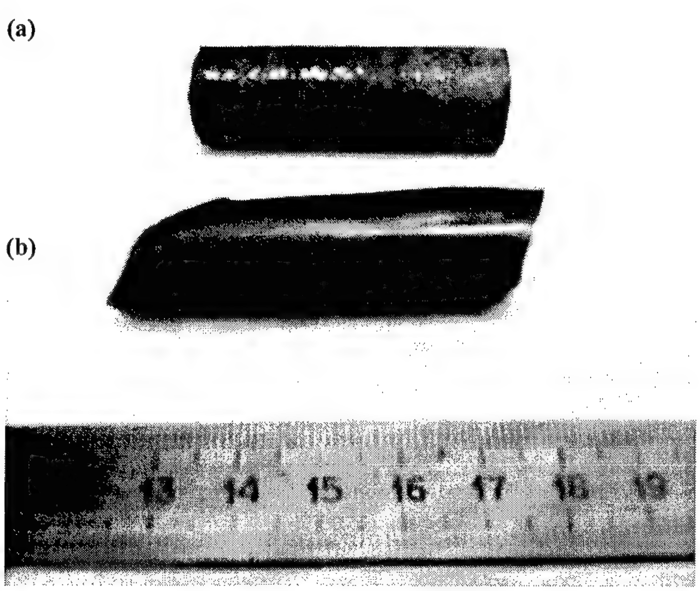


Fig. 8. OM (a) and TEM (b) micrographs of the W in the as- received state (transverse section).

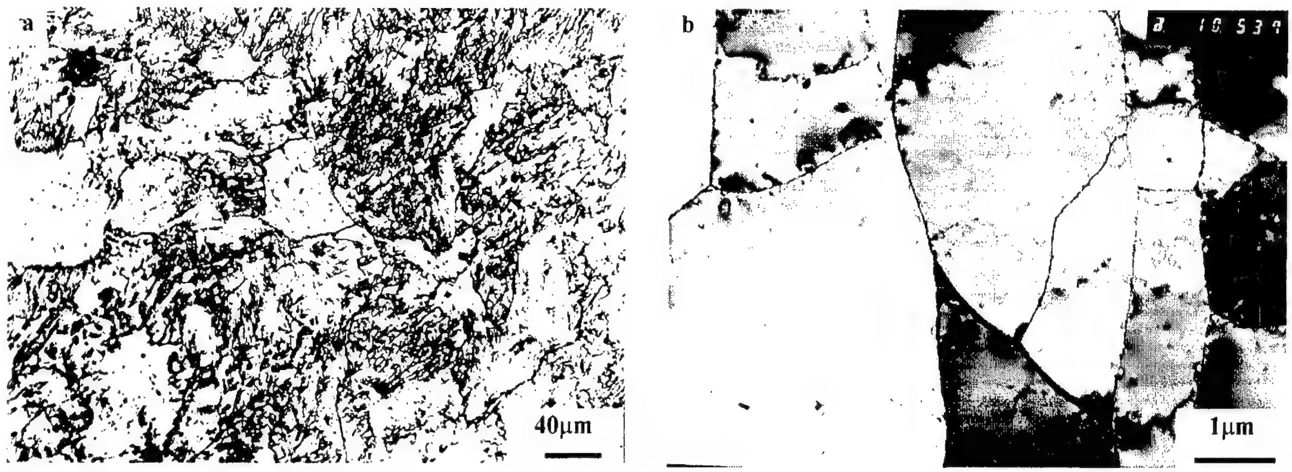



Fig. 9. OM micrographs of ECAP processed W after 4 passes in transverse (a) and longitudinal (b) sections, and after 8 passes in transverse (c) and longitudinal (d) sections.

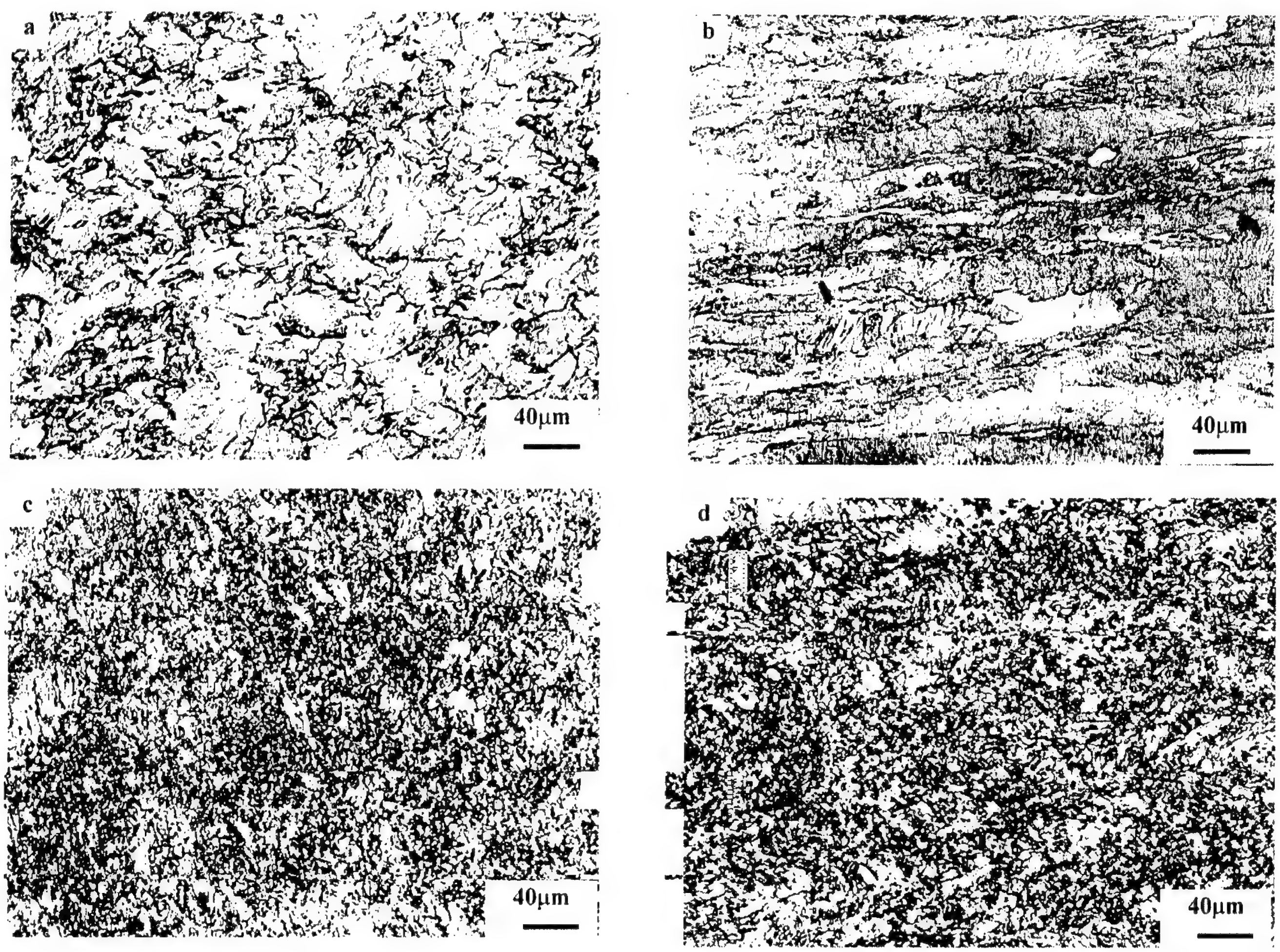


Fig. 10. TEM micrographs and SAED patterns of ECAP processed W after 4 passes in transverse (a) and longitudinal (b) sections, and after 8 passes in transverse (c) and longitudinal (d) sections.

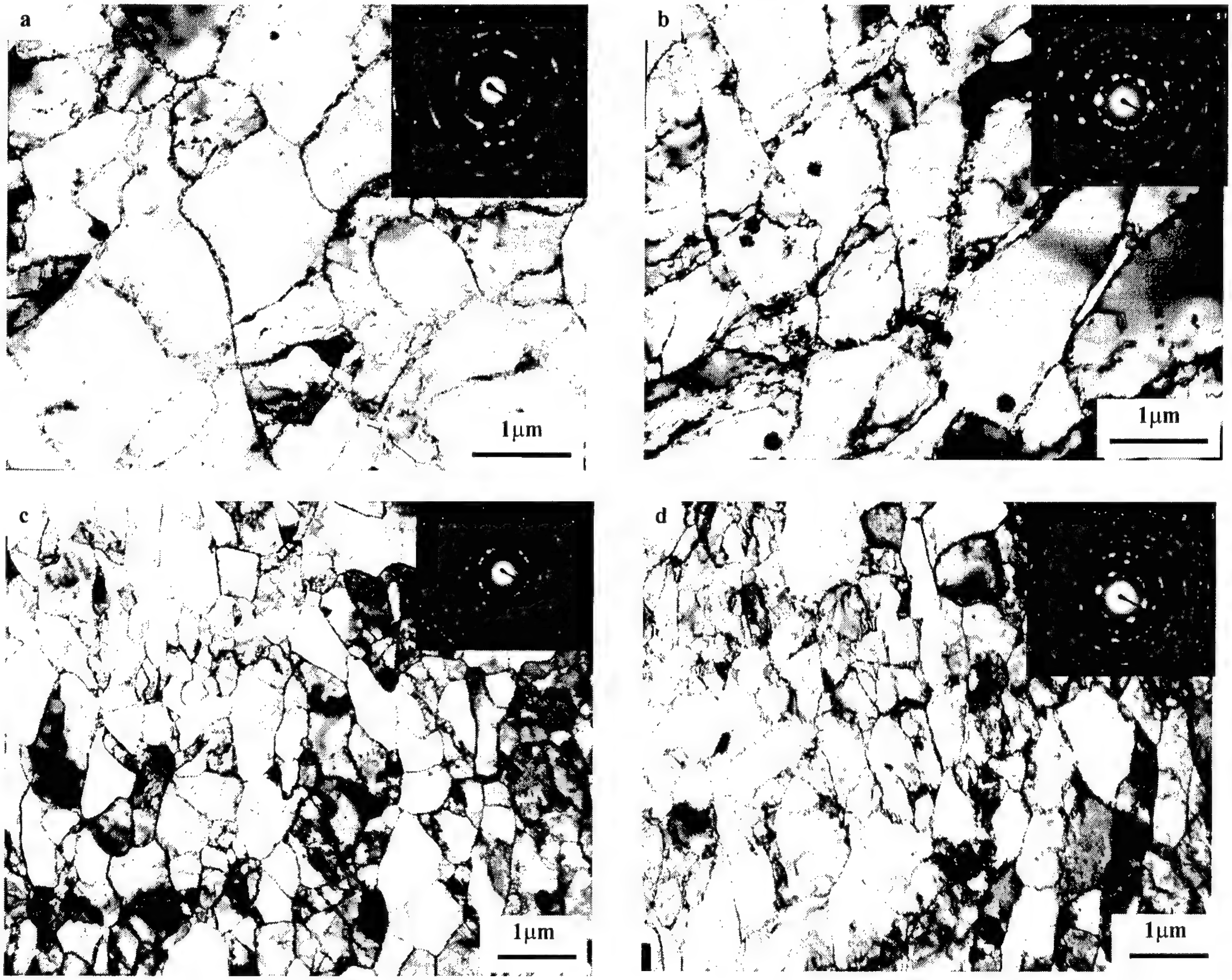



Fig. 11. TEM micrographs and SAED of HPT strained W: bright (a) and dark (b) field images.

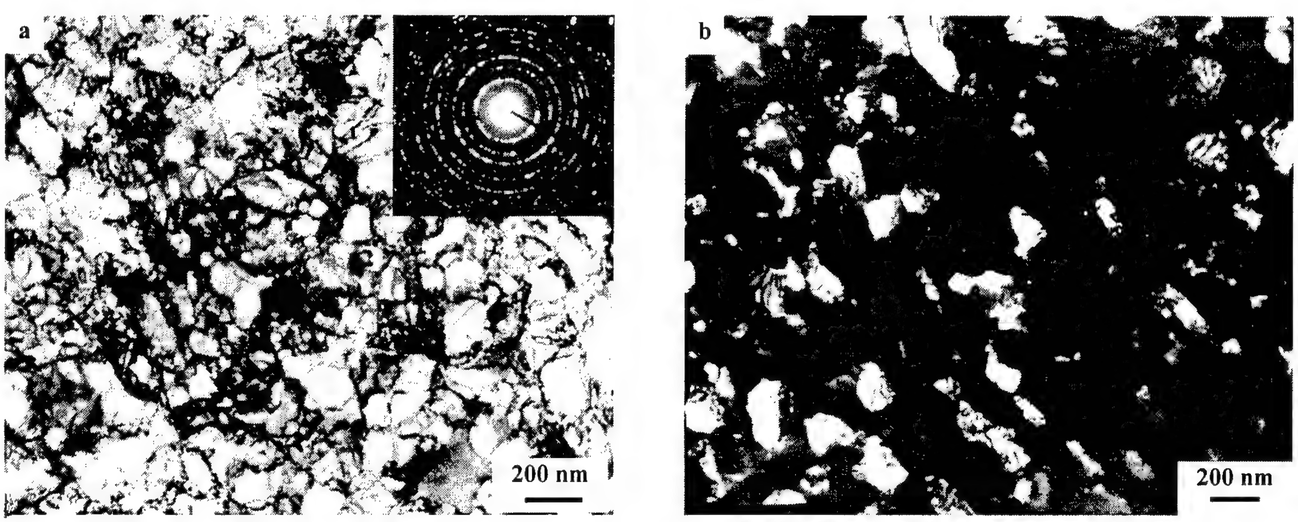


Table 1. SPD W. Crystallite size and elastic microdistortions in the <110> direction.

| | Initial state | HPT straining | ECA pressing |
|---|---|---|---|
| Crystallite size, nm | 416±31 | 95±5 | 91±8 |
| Microdistortions, % | 0.056±0.002 | 0.249±0.005 | 0.172±0.003 |

Fig. 12. TEM micrographs and SAED patterns of ECAP processed W after 4 passes in transverse (a) and longitudinal (b) sections, and after 8 passes in transverse (c) and longitudinal (d) sections.

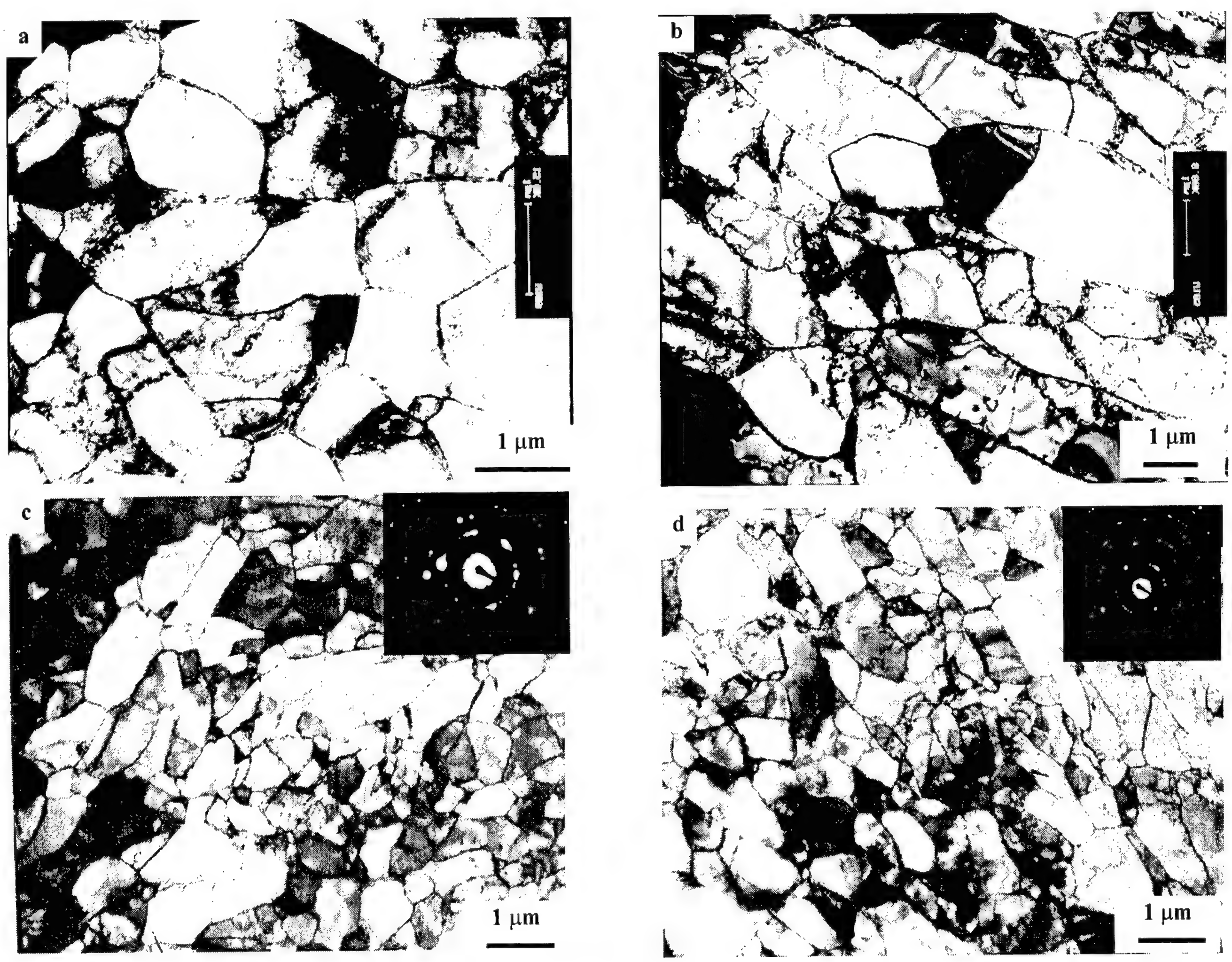
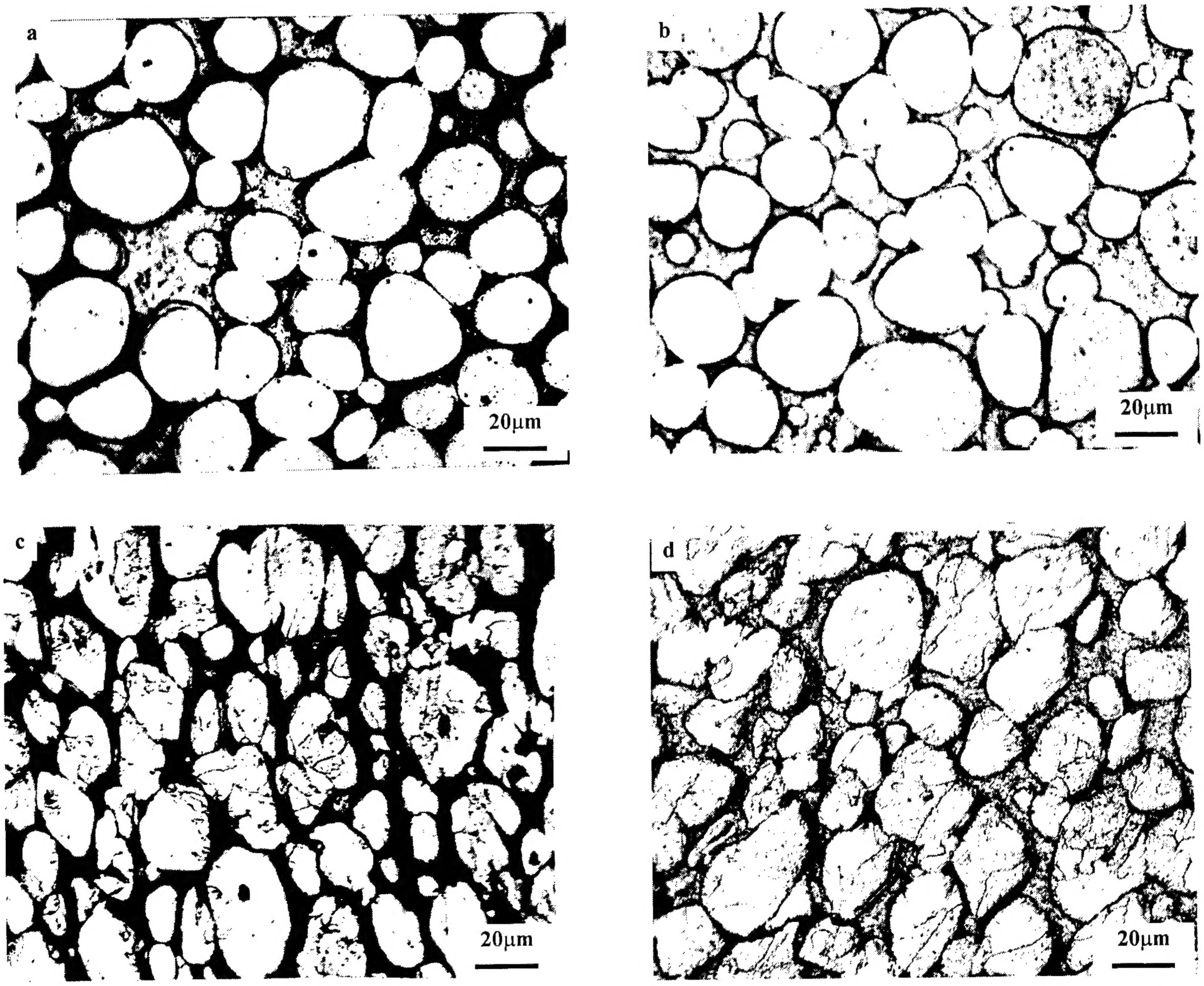


Fig. 13. OM micrographs of W heavy alloy before (a), (b) and after ECAP (c), (d) in transverse and longitudinal sections of billet



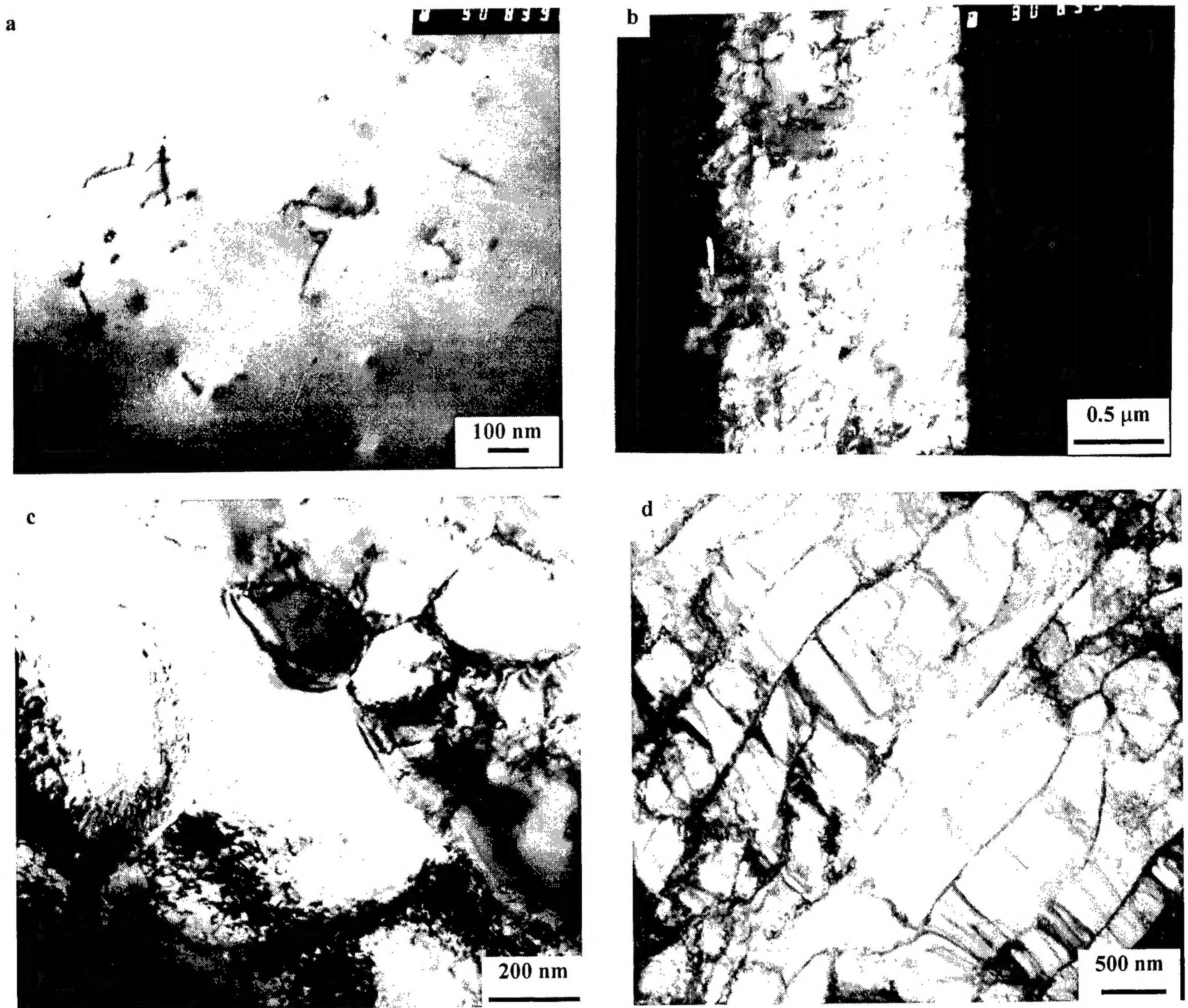



Fig. 14. TEM micrographs of W heavy alloy before (a), (b) and after ECAP in transverse (c), in longitudinal (d) sections of a billet.

Fig. 15. Effect of the number passes of ECAP processed W on TEM (sub)grain size (a) and microhardness (b) in transverse section of billets.

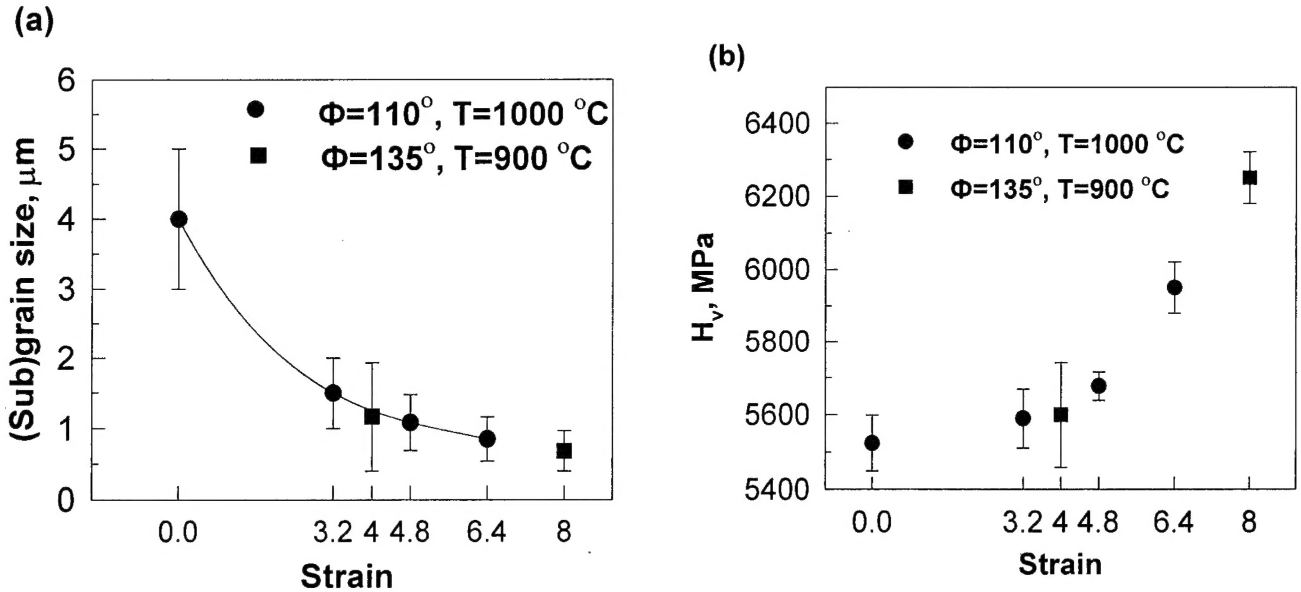



Fig. 16. The influence of the testing temperature on the mechanical properties of W before and after ECAP.

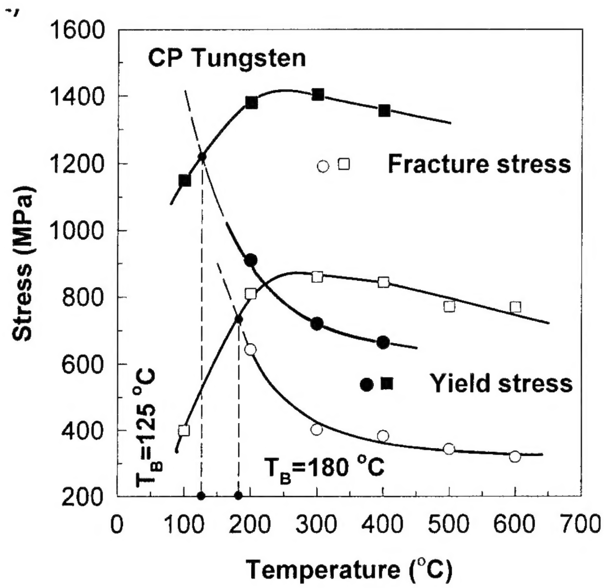


Fig. 17. The effects of structural states of W on fracture surface: initial state (a), ECAP (b) and HPT straining (c) and (d).